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(54) Resin composition for electric insulation materials for vehicles.

(57) The present invention is directed to a resin composition for electric insulation materials for vehicles such as distributor caps which is prepared by blending a crystalline propylene-ethylene copolymer resin with 3 to 10 wt.% of a glass fiber, 10 to 20 wt.% of talc or mica, 5 to 20 wt.% of silica, 4 to 20 wt.% of modified polyolefin resins, 10 to 30 wt.% of halogen-containing flame retardants and 3 to 15 wt.% of flame retardant auxiliaries. In the resin composition of the present invention, balance is kept in all points of mechanical characteristics, mechanical characteristics at high temperature, electrical characteristics, nitric acid resistance, weld characteristics, blooming resistance and flame retardance.

Resin Composition for electric insulation materials for vehicles

CROSS-REFERENCE TO RELATED APPLICATION

The present invention claims a priority on the basis of Japanese Patent Application No. 298,019/1986.

BACKGROUND OF THE INVENTION

(1) Field of the Invention

The present invention relates to a resin composition applied to electric insulation materials for vehicles. More specifically, it relates to a resin composition for distributor caps which is prepared by blending a crystalline propylene-ethylene copolymer resin with glass fibers, talc or mica, silica, modified polyolefin resins, halogen-containing flame retardants and flame retardant auxiliaries.

(2) Description of the Prior Art

Heretofore, as materials for distributor caps have been used, for example, an epoxy resin, a polybutylene terephthalate resin containing glass fibers (hereafter referred to as the glass fiber-containing PBT), a polypropylene resin containing talc and a polypropylene resin containing talc and flame retardants.

The electric insulation materials for vehicles in this invention mean the electric insulation materials for motor vehicles, used at high temperature, under often wetly contaminated circumstances, like engine room, and at high voltage. It typically means the material for distributor caps.

The epoxy resins have the longest history and have been used in extensive fields, but their product cost is high because of a high material cost and manufacturing cost, and they are poor in impact resistance and therefore are liable to break during transportation disadvantageously. The glass fiber-containing PBT and the talc-containing polypropylene resin can solve the above problems. However, the glass fiber-containing PBT has a poor tracking resistance, and therefore when distributor caps comprising the glass fiber-containing PBT is used in a briny air district, the outer surfaces of the distributor caps are partially carbonized, so that the problem of bad insulation occurs. As for the talc-containing polypropylene resin, mechanical characteristics (stiffness) at high temperature and weld characteristics are poor. Furthermore, a common problem of the glass fiber-containing PBT and the talc-containing polypropylene is that flame retardance is poor.

Here, the reason why the tracking resistance is necessary is as follows: When the distributor caps are under circumstances where the surfaces of them are liable to be brought into contact with a saline solution or the like, as in a briny air district, the surfaces are wetly contaminated and further partially carbonized by a minute electric discharge so as to allow an electrical conduction therethrough, so that their insulation properties become bad.

The tracking resistance is due to the molecular structure of the resin material. A resin such as the PBT which has a benzene ring in a molecule is bad in tracking resistance. This reason is as follows: Carbon bonds in the benzene ring are firm, and therefore the resin is hard to gasify by a heat energy of the minute discharge, so that conductive free carbons having a graphite structure are apt to be left on the surfaces. To the contrary, the polypropylene resin has no benzene ring in the molecule, and therefore its tracking resistance is extremely good. Accordingly, from the viewpoint of the tracking resistance, the polypropylene resin is most desirable as a material for distributor caps.

The reason why the above-mentioned mechanical characteristics (stiffness) at high temperature is necessary is as follows: In recent years, conditions in the vicinity of an automotive engine are stern, and with regard to the distributor cap under such conditions, a higher heat resistance is required. However, the talc-containing polypropylene resin has poor mechanical characteristics (stiffness) at high temperature (e.g., such temperature as to cause a thermal distortion under a fiber stress of 18.6kg/cm²: usually 90 to 100°C). Therefore, when a tower portion of the cap onto which a load from a high-tension cord is applied is vibrated at a high temperature, this portion tends to be deformed and damaged.

The above-mentioned problems of the weld characteristics include a problem of a bad weld appearance which looks as if cracks occur in the molded article of the distributor caps, and another serious problem that

cracks take place in electrode portions of the caps and a weld portion on the periphery of insert fittings. That is, such cracks in the weld portion are caused by a progressive crystallization subsequent to molding, by a post shrinkage and by a heat stress generated from cooling/heating cycle in the case that the caps are exposed to high-temperature conditions in an engine room of an automobile for a long period of time, or in the case that it is exposed to rapid temperature change conditions in winter.

Further, the reason why the above-mentioned flame retardance is necessary is as follows; when a connector for distribution is fixed improperly, an arc discharge is generated during driving an automobile and the tower portion of a distributor cap is carbonized by a discharge energy of this arc discharge. When the carbonization progresses, the cap must be changed with a new one.

The polypropylene resin containing talc and a flame retardant can eliminate the drawback of the poor flame retardance which the glass fiber-containing PBT and the talc-containing polypropylene resin have. With regard to the polypropylene resin containing talc and the flame retardant, however, as a result of the incorporation of the flame retardant, the mechanical characteristics (tensile strength) decline, and in addition, the poor weld characteristics which are the drawback of the talc-containing polypropylene resin are worsened. Furthermore, when this resin is used under high-temperature condition, the used flame retardant blooms to impair its appearance and a flame retardance declines disadvantageously.

In view of the above-mentioned conventional technical problems, the object of the present invention is to provide a resin composition suitable for electric insulation materials for vehicles in which balance is kept in all points of cost, workability, impact strength, mechanical characteristics (tensile strength), mechanical characteristics (stiffness) at high temperature, tracking resistance, weld characteristics and flame retardance.

SUMMARY OF THE INVENTION

The present invention is directed to a resin composition usable for electric insulation materials for vehicles such as distributor caps which is prepared by blending a crystalline propylene-ethylene copolymer resin with glass fibers, talc or mica, silica, modified polyolefin resins, halogen-containing flame retardants and flame retardant auxiliaries in a suitable ratio, and in the resin composition, balance is kept in all points of mechanical characteristics, mechanical characteristics at high temperature, electrical characteristics, nitric acid resistance, weld characteristics, blooming resistance and flame retardance.

DESCRIPTION OF THE SPECIFIC EMBODIMENT

With regard to the five problems of cost, workability, impact strength, mechanical characteristics (tensile strength) and tracking resistance, they have already been solved by a conventional talc-containing polypropylene resin.

Flame retardance can be obtained by adding flame retardants in accordance with a known technique, but when the flame retardants are added directly to the talc-containing polypropylene resin, its mechanical characteristics (tensile strength) are reduced, and weld characteristics which have been originally poor is further worsened inconveniently. Therefore, points of the development are to add the flame retardant without sacrificing any mechanical characteristics (tensile strength) and to improve the mechanical characteristics (stiffness) at high temperature and the weld characteristics up to a higher level than in the talc-containing polypropylene resin. That is, kinds and optimum composition ratios of inorganic fillers as reinforcing materials and the flame retardant are crucial to the success in the present invention.

Further, it is attributable to a morphology of a talc grain crystal that the talc-containing polypropylene resin is poor in mechanical characteristics (stiffness) at high temperature and weld characteristics. That is, the talc is a lamellar crystal alike mica and thus is oriented in the form of a layer in the weld portion of a molded article, so that a tensile strength of this weld portion is as poor as 20 to 40% of the portion other than the weld portion. This is the cause of the poor weld characteristics. Moreover, the talc has not so a high aspect ratio which has a great influence on the strength and stiffness of the molded article comprising an inorganic filler composite resin composition, and this is the cause of lowering the mechanical characteristics (stiffness) at high temperature. In consequence, for the purpose of improving the mechanical characteristics (stiffness) at high temperature and the weld characteristics, the morphology of the inorganic fillers which will be blended with the polypropylene resin also is an important factor.

When it is investigated what inorganic fillers should be blended, nitric acid resistance and arc resistance must be additionally taken into consideration in compliance with circumstances where the distributor caps

are used, besides the above-mentioned mechanical characteristics (stiffness) at high temperature and the weld characteristics. The reason why the nitric acid resistance is necessary is as follows: A nitrogen compound (NO_x), which is generated during the use of the distributor, reacts with water in air present in the distributor to produce nitric acid. And an inorganic matter, if any, in the distributor cap material reacts with the thus produced nitric acid in order to form nitrates (e.g., calcium nitrate and magnesium nitrate), and these nitrates are deposited on the surface of the distributor cap. Since the nitrates are deliquescent, the distributor cap loses electrical insulation properties. This is the reason why the nitric acid resistance is necessary.

The reason why the arc resistance is necessary is as follows: The distributor cap, when used, is sometimes exposed to an arc discharge which is generated between electrodes, so that an intensive discharge energy is thereby generated to provide high temperature, which attacks the outer surface of the cap locally. As a result, gasification and carbonization make slow progress, and at last a carbon track (a conductive path of carbon) is formed by the carbonization in order to lead to a bad insulation. This is the reason why the arc resistance is necessary. Such a requirement is closely connected with the above-mentioned resistance to the tracking phenomenon which tends to occur on the cap surface in the wet contamination state. Accordingly, the polypropylene resin which is liable to gasify by the discharge energy is basically excellent in arc resistance, but nevertheless even in the same kind of resin, this arc resistance varies with kinds and amounts of fillers. For example, if a fibrous filler such as a glass fiber is added in large quantities, the arc resistance will deteriorate.

In the first place, the inventors of the present application have investigated and evaluated various fillers in accordance with the morphology from the four viewpoints of the mechanical characteristics (stiffness) at high temperature, the weld characteristics, the nitric acid resistance and the arc resistance. The evaluation is carried out by employing talc as standards (Table 1).

As understood from Table 1, any filler singly did not satisfy all of the four characteristics, and it was made clear that only one kind of filler could hardly achieve the above object. Then, the present inventors chose fillers having different characteristics (the mechanical characteristics at high temperature, the weld characteristics and the arc resistance) from the fillers which meets the nitric acid resistance, and multi-component systems, each comprising two or more kinds of fillers were investigated. As a result, it has been found that a blend of talc or mica as a lamellar filler, a glass fiber as a fibrous filler and silica as an amorphous filler has the most balanced characteristics.

With regard to the provision of the flame retardance, it is a known technique to add the flame retardant to the polypropylene resin composition. However, when a resin composition which has been prepared merely by adding a kind of halogen-containing flame retardant to the polypropylene resin composition is used to form an article, and when the latter article is used under high-temperature conditions for a certain period of time, the used flame retardant bleeds on the surface of the article, so that the appearance of the article is worsened and simultaneously the flame retardance is lowered disadvantageously. Therefore, in order to solve this problem of the poor blooming resistance, the polypropylene resin having a high compatibility with the flame retardant is chosen, and there is employed the notion that the flame retardant having a heat resistance and the nitric acid resistance is effective. As a result of various researches, the following fact has been found: When there were used a crystalline propylene-ethylene copolymer resin, a halogen-containing flame retardant having a melting point of 200°C or more [e.g., decabromodiphenyl ether, dodecachlorododecahydridimethanodibenzocyclooctene (trade name Dechloranplus)] and an antimony compound (e.g., antimony trioxide) or a boron compound (e.g., zinc borate or borax) as a flame retardant auxiliary, the drawbacks of the bad appearance due to the bleed and the decline in the flame retardance can be mitigated remarkably.

On the basis of the above-mentioned fundamental investigation result, detailed researches have been further conducted intensively, and as a result, it has been found that a compound having the undermentioned constitution can achieve the object of the present invention.

The present invention constitutes a resin composition for electric insulation material for vehicles such as distributor caps which is prepared by blending a crystalline propylene-ethylene copolymer resin with 3 to 10 wt.% of glass fibers, 10 to 20 wt.% of talc or mica, 5 to 20 wt.% of silica, 4 to 20 wt.% of modified polyolefin resins, 10 to 30 wt.% of halogen-containing flame retardants and 3 to 15 wt.% of flame retardant auxiliaries, said percentages being based on the resin composition.

The resin composition in the present invention is the most appropriately used for distributor caps which are liable to be brought into high temperature and wetly contaminated circumstances and are loaded high voltage and are brought into contact with spark, among the electric insulation materials for vehicles.

The crystalline propylene-ethylene copolymer resin used in the present invention is composed of 2 to 35 wt.% of ethylene and has a melt flow rate (MFR; a discharge of a melted resin for 10 minutes in the

case that 2.16kg is loaded at temperature of 230°C) of 1.0 to 80g/10 minutes, and in particular, a crystalline propylene-ethylene block copolymer resin is preferable.

With regard to the glass fibers used in the present invention, its diameter and length are not particularly limited, and the commercially available glass fibers for thermoplastic resins may usually be used. An amount of the glass fibers to be blended is within the range of 3 to 10 wt.%, preferably 5 to 8 wt.%. When the content of the glass fiber is less than 3 wt.%, the electric insulation material such as a distributor cap obtained by molding its resin composition has poor mechanical characteristics (stiffness) at high temperature unpreferably. Inversely, when it is more than 10 wt.%, the electric insulation materials such as a distributor cap obtained by molding its resin composition has excellent mechanical characteristics (stiffness) at high temperature but simultaneously has poor arc characteristics unpreferably.

With regard to talc or mica used in the present invention, its grain diameter is not particularly limited and a commercial talc or mica, desirably having a great aspect ratio, ordinarily used for thermoplastic resins can be used. The talc or mica may be employed without any treatment, but for the purpose of improving adhesive properties with a matrix resin or dispersion properties, grains of this material may be coated, prior to its use, with various organic titanate coupling agents, silane coupling agents, fatty acids, metallic salts of fatty acids or fatty esters. An amount of the talc or mica to be blended is within the range of 10 to 20 wt.%, based on the weight of the resin composition. When the content of the talc or mica is less than 10 wt.%, the electric insulation materials such as a distributor cap obtained by molding its resin composition has insufficient mechanical characteristics (stiffness) at high temperature unpreferably, and when it is in excess of 20 wt.%, articles obtained by molding its resin composition have poor weld characteristics unpreferably.

With regard to the silica used in the present invention, its grain diameter is not particularly limited, and the commercial silica for thermoplastic resins may be usually used. The silica may be employed without any treatment, but for the purpose of improving adhesive properties with a matrix resin or dispersion properties, grains of the silica may be coated, prior to its use, with various organic titanate coupling agents, silane coupling agents, fatty acids, metallic salts of fatty acids or fatty esters, in common with the above-mentioned talc or mica. An amount of the silica to be blended is within the range of 5 to 20 wt.%, preferably 5 to 15 wt.%, based on the weight of the resin composition. When the content of the silica is less than 5 wt.%, articles obtained by molding its resin composition have a poor mechanical characteristics (stiffness) at high temperature unpreferably. Further, when it is in excess of 20 wt.%, a granulation in the manufacturing process of composition pellets cannot be carried out smoothly.

The modified polyolefin resin used in the present invention can be obtained by melting and kneading a polyolefin resin such as a polyethylene, a propylene homopolymer, a propylene-ethylene copolymer or a polybutene together with an unsaturated carboxylic acid or its anhydride (e.g., maleic anhydride) in the presence of an organic peroxide. Concrete examples of the usable modified polyolefin resins include modified polyethylenes, modified propylene homopolymers, modified propylene-ethylene copolymers and modified polybutenes, the modified propylene-ethylene copolymers particularly preferred. An amount of the modified polyolefin resin to be blended is within the range of 4 to 20 wt.%, preferably 6 to 12 wt.%, based on the weight of the resin composition.

The halogen-containing flame retardants used in the present invention have a melting point of 200°C or more, and concrete examples of the usable halogen-containing flame retardants include decabromodiphenyl ether, dodecachlorododecahydridimethanodibenzocyclooctene (trade name Dechloran-plus) and mixtures thereof. An amount of the halogen-containing flame retardants to be blended is within the range of 10 to 30 wt.%, preferably 10 to 20 wt.%, based on the weight of the resin composition. When the content of the halogen-containing flame retardants is less than 10 wt.%, articles obtained by molding its resin composition cannot have such a high flame retardance as to be expected, unpreferably. Inversely, when it is more than 30 wt.%, articles obtained by molding its resin composition are poor in mechanical characteristics (tensile strength), and a granulation in the manufacturing process of composition pellets cannot be carried out smoothly.

Concrete examples of the flame retardant auxiliaries used in the present invention include an antimony compound such as antimony trioxide and boron compounds such as zinc borate and borax. An amount of the flame retardant auxiliary to be blended is preferably in a weight ratio of 1/4 to 1/2 based on the weight of the halogen-containing flame retardant.

In addition to the above-mentioned blending materials, the following materials may be blended with the resin composition for electric insulation materials for vehicles regarding the present invention: Colorants such as dyes or pigments, nucleating agents, lubricants, anti-oxidants, heat stabilizers, light stabilizers, release agents, crosslinking agents, radical generators, foaming agents and the like.

A manufacturing process of the resin composition for distributor caps regarding the present invention is

not particularly limited, and the following process is one example: In a stirring mixer such as a Henschel mixer (trade name) or a supermixer are placed the above-mentioned crystalline propylene-ethylene copolymer resin and specific amounts of talc or mica, silica, modified polyolefin resins, halogen-containing flame retardants and flame retardant auxiliaries, and stirring is then carried out for a period of 30 seconds to 3 minutes. To the resulting mixture, glass fibers are added, and in a stirring mixer such as a tumble mixer, the mixture is then rotated 5 to 20 times for the sake of stirring/mixing. The resulting mixture is melted, kneaded and pelletized by the use of a Banbury mixer, rolls and an extruder. A temperature for melting and kneading is within the range of 180° to 280°C, preferably 200 to 250°C.

10 EXAMPLES

Now, the present invention will be described in detail in reference to examples, but it should not be limited by these examples.

15 Various tests in the examples and comparative examples were each performed by the following procedure:

20 (1) Mechanical Characteristics:

Mechanical characteristics were evaluated by measuring a tensile strength (in accordance with JIS K-7113).

25 (2) Mechanical Characteristics at High Temperature (stiffness):

Mechanical characteristics at high temperature (stiffness) were evaluated by measuring a heat distortion temperature (fiber stress of 18.6 kg/cm²) (in accordance with JIS K-7207).

30 (3) Electrical Characteristics:

Electrical characteristics were evaluated by measuring arc resistance (in accordance with ASTM D-495).

35 (4) Nitric Acid Resistance:

Each specimen having a size of 50×50×2 mm which was prepared by injection molding was immersed into a commercial nitric acid (concentration 61%; specific gravity 1.38), and the generation of gas bubbles was observed. The nitric acid resistance was evaluated by estimating the specimen of no gas bubbles to be good and the specimen of the bubble generation to be bad.

40 (5) Weld Characteristics:

A weld strength was evaluated by measuring a weld tensile strength (in accordance with JIS K-7113), and weld characteristics were evaluated by visually observing a weld portion of a JIS No. 1 type weld tensile dumbbell specimen which is prepared by injection molding (in which a weld was formed in the middle of JIS No. 1 type tensile dumbbell specimen, L = 178 mm). In this case, the specimen on which the weld portion was noticeably perceptible was estimated to be bad, and the specimen on which the weld portion was scarcely perceptible was estimated to be good.

45 (6) Blooming Resistance:

Each specimen having a size of 50×50×2 mm prepared by injection molding was allowed to stand at 100°C for 240 hours in an oven, and afterward its appearance was observed visually. In this case, the bloomed specimen was estimated to be bad, and the non-blooming specimen was estimated to be good.

(7) Flame Retardance:

Each specimen having a size of 127×12.7×1.6 mm which was prepared by injection molding was subjected to a burning test in accordance with a vertical burning test process of American UL Standard Subject 94 (UL 94), whereby a flame retardant was evaluated.

Examples 1 to 17 and Comparative Examples 1 to 18:

In Examples 1 to 17, in a Henschel mixer (trade name) were placed a propylene-ethylene block copolymer resin having a melt flow rate of 10 g/10 minutes and containing 8.5 wt.% of ethylene, talc or mica, silica, a modified propylene-ethylene copolymer resin, decabromodiphenyl ether and antimony trioxide in predetermined amounts, as set forth in Tables 2 and 3, and stirring/mixing was then carried out for 1 minute. A predetermined amount of glass fibers was afterward added to the resulting mixture, and the mixture was rotated 10 times in a tumbler for the sake of stirring/mixing. The resulting mixture was melted, kneaded and pelletized at a melting/kneading temperature of 230°C by the use of a twin-screw extruder having a bore diameter of 30 mm.

In Comparative Examples 1 to 18, as set forth in Tables 4 and 5 given below, predetermined amounts of components to be blended were stirred and mixed in a Henschel mixer (trade name) and a tumbler, and the resulting mixture was then melted, kneaded and pelletized by the use of a twin-screw extruder having a bore diameter of 30 mm.

Specimens having a predetermined shape were molded at a resin temperature of 250°C and at a mold temperature of 50°C from the thus prepared pellets by injection molding in the respective examples and comparative examples, and the evaluation of these specimens was made with regard to mechanical characteristics, mechanical characteristics at high temperature, electrical characteristics, nitric acid resistance, weld characteristics, blooming resistance and flame retardance. The results of the evaluation are summarized in Table 2 to 5.

As be definite from Tables 2 and 3, the molded articles of the resin compositions regarding the present invention in Examples 1 to 17 were sufficiently balanced in points of mechanical characteristics (tensile strength), mechanical characteristics (stiffness) (which were represented by the heat distortion temperature) at high temperature, electrical characteristics (arc resistance), nitric acid resistance, weld characteristics, blooming resistance and flame retardance. In particular, as be apparent from the comparison between Examples 1 to 17 and Comparative Examples 1 to 8 in which the amounts of talc or mica were in excess of 20 wt.%, the poor weld characteristics which were the drawback of the talc-containing polypropylene could be solved by adding the silica and the glass fiber.

Further, as be apparent from the comparison between Examples 1 to 17 and Comparative Examples 2,3,4,7,10,16 and 18 in which the basic polymer was the crystalline propylene homopolymer resin, the blooming phenomenon (a powdery coating phenomenon on the surface of the molded article under high-temperature conditions) which was the drawback of the halogen-containing flame retardants (decabromodiphenyl ether) used herein could be solved by using the propylene-ethylene block copolymer resin. In Comparative Examples 1,2,3,4,5,9,10 and 11 in which amounts of the glass fibers were less than 3 wt.%, the mechanical characteristics (stiffness) at high temperature were poor. In Comparative Examples 12,13 and 14 in which amounts of the glass fibers were more than 10 wt.%, the electrical characteristics (arc resistance) were insufficient. In Comparative Examples 1,3,6,9,12 and 15 in which amounts of the halogen-containing flame retardants were less than 10 wt.%, the flame retardant was poor. In Comparative Examples 15, 16,17 and 18 in which calcium carbonate was used, the nitric acid resistance was bad.

To sum up, when the resin composition of the present invention is used, the distributor caps can be obtained which are excellent in mechanical characteristics, mechanical characteristics at high temperature, electrical characteristics, nitric acid resistance, weld characteristics, blooming resistance and flame retardance.

Table 1

Evaluation of Each Inorganic Filler to Characteristics

<u>Morphology</u>	<u>Filler</u>	<u>Mechanical Charac. (stiffness) at High Temp.</u>	<u>Weld Charac.</u>	<u>Nitric Acid Resistance</u>	<u>Arc Resistance</u>
Lamellar	Talc	Δ	X	O	O
	Mica	O	X	O	O
Fibrous	Calcium Silicate	Δ	Δ	X	Δ
	Glass Fiber	O	O	O	X
Amorphous	Calcium Carbonate	X	O	X	O
	Barium Sulfate	X	O	O	O
	Silica	Δ	O	O	O
	Glass Beads	X	O	O	X
	Magnesium Hydroxide	X	O	X	O
	Aluminum Hydroxide	X	O	X	O

O: Good, Δ: Middle, X: Bad

Table 2 (I)

	<u>Ex. 1</u>	<u>Ex. 2</u>	<u>Ex. 3</u>	<u>Ex. 4</u>	<u>Ex. 5</u>	<u>Ex. 6</u>
Blending Materials (wt.%)						
PP Block Copolymer	54	49	44	54	44	47
Glass Fiber	5	5	5	5	5	5
Talc	10	15	10	10	10	10
Mica	0	0	10	0	10	0
Silica	10	10	10	5	5	5
Modified PP	6	6	6	6	6	6
DBDE	10	10	10	15	15	20
Antimony Trioxide	5	5	5	5	5	7

PP Block Copolymer: Propylene-ethylene block copolymer resin having a melt flow rate of 10 g/10 minutes and containing 8.5 wt.% of ethylene.

Modified PP: Maleic anhydride-modified propylene-ethylene copolymer resin.

DBDE: Decabromodiphenyl ether.

Table 2 (II)

Blending Materials (wt.%)	<u>Ex. 7</u>	<u>Ex. 8</u>	<u>Ex. 9</u>	<u>Ex. 10</u>	<u>Ex. 11</u>	<u>Ex. 12</u>
PP Block Copolymer	56.5	51.5	51.5	51.5	46.5	41.5
Glass Fiber	7.5	7.5	7.5	7.5	7.5	7.5
Talc	10	5	10	10	10	15
Mica	0	10	0	0	10	0
Silica	5	5	5	10	5	10
Modified PP	6	6	6	6	6	6
DBDE	10	10	15	10	10	15
Antimony Trioxide	5	5	5	5	5	5

Table 2 (III)

	<u>Ex. 1</u>	<u>Ex. 2</u>	<u>Ex. 3</u>	<u>Ex. 4</u>	<u>Ex. 5</u>	<u>Ex. 6</u>
Tensile Strength (kg/cm ²)	360	340	330	380	320	330
Heat Distortion Temp. (18.6 kg/cm ² ; °C)	115	115	120	105	120	125
Arc Resistance (sec)	100	100	100	90	90	80
Nitric Acid Resistance	Good	Good	Good	Good	Good	Good
Weld Characteristics Tensile Strength (kg/cm ²)	180	170	145	190	140	165
Appearance	Good	Good	Good	Good	Good	Good
Blooming Resistance (100°C; 240 hrs)	Good	Good	Good	Good	Good	Good
Flame Retardance (UL94; Vertical Burning Test; 1.6t; Class)	V-2	V-2	V-2	V-1	V-0	V-0

Table 2 (IV)

	<u>Ex. 7</u>	<u>Ex. 8</u>	<u>Ex. 9</u>	<u>Ex. 10</u>	<u>Ex. 11</u>	<u>Ex. 12</u>
Tensile Strength (kg/cm ²)	420	410	400	400	400	400
Heat Distortion Temp. (18.6 kg/cm ² ; °C)	120	125	125	120	125	125
Arc Resistance (sec)	100	100	90	100	100	90
Nitric Acid Resistance	Good	Good	Good	Good	Good	Good
Weld Characteristics Tensile Strength (kg/cm ²)	200	190	190	200	160	200
Appearance	Good	Good	Good	Good	Good	Good
Blooming Resistance (100°C; 240 hrs)	Good	Good	Good	Good	Good	Good
Flame Retardance (UL94; Vertical Burning Test; 1.6t; Class 2)	V-2	V-2	V-1	V-2	V-2	V-1

Table 3 (I)

	<u>Ex. 13</u>	<u>Ex. 14</u>	<u>Ex. 15</u>	<u>Ex. 16</u>	<u>Ex. 17</u>
Blending Materials (wt.%)					
PP Block Copolymer	39.5	36.5	36.5	54	49
Glass Fiber	7.5	7.5	7.5	10	10
Talc	10	20	10	10	10
Mica	0	0	10	0	0
Silica	10	10	10	5	5
Modified PP	6	6	6	6	6
DBDE	20	15	15	10	15
Antimony Trioxide	7	5	5	5	5

PP Block Copolymer: Propylene-ethylene block copolymer resin having a melt flow rate of 10 g/10 minutes and containing 8.5 wt.% of ethylene.

Modified PP: Maleic anhydride-modified propylene-ethylene copolymer resin.

DBDE: Decabromodiphenyl ether.

Table 3 (II)

	<u>Ex. 13</u>	<u>Ex. 14</u>	<u>Ex. 15</u>	<u>Ex. 16</u>	<u>Ex. 17</u>
Tensile Strength (kg/cm ²)	350	400	400	450	430
Heat Distortion Temp. (18.6 kg/cm ² ; °C)	122	125	125	130	130
Arc Resistance (sec)	80	90	90	100	90
Nitric Acid Resistance	Good	Good	Good	Good	Good
Weld Characteristics Tensile Strength (kg/cm ²)	170	160	150	200	210
Appearance	Good	Good	Good	Good	Good
Blooming Resistance (100°C; 240 hrs)	Good	Good	Good	Good	Good
Flame Retardance (UL94; Vertical Burning Test; 1.6t; Class)	V-0	V-1	V-1	V-2	V-1

Table 4 (I)

Blending Materials (wt.%)	1	2	Comparative Example		
			3	4	5
PP Homopolymer	60	45	60	55	0
PP Block Copolymer	0	0	0	0	55
Glass Fiber	0	0	0	0	0
Talc/Mica	40/0	40/0	30/0	10/20	30/0
Silica	0	0	0	0	0
Modified PP	0	0	0	0	0
DBDE	0	10	7	10	10
Antimony Trioxide	0	5	3	5	5

PP Block Copolymer: Propylene-ethylene block copolymer resin having a melt flow rate of 10 g/10 minutes and containing 8.5 wt.% of ethylene.

Modified PP: Maleic anhydride-modified propylene-ethylene copolymer resin.

DBDE: Decabromodiphenyl ether.

Table 4 (II)

Blending Materials (wt%)	7	8	Comparative Example			11	12
			9	10			
PP Homopolymer	44	0	60	45		0	49
PP Block Copolymer	0	44	0	0		45	0
Glass Fiber	5	5	0	0		0	15
Talc/Mica	10/20	10/20	15/0	15/0		15/0	10/10
Silica	0	0	25	25		25	10
Modified PP	6	6	0	0		0	6
DBDE	10	10	0	10		10	0
Antimony Trioxide	5	5	0	5		5	0

Table 4 (III)

	<u>1</u>	<u>2</u>	<u>Comparative Example</u>		<u>5</u>	<u>6</u>
			<u>3</u>	<u>4</u>		
Tensile Strength (kg/cm ²)	350	320	330	320	300	450
Heat Distortion Temp. (18.6 kg/cm ² ; °C)	95	100	100	105	95	130
Arc Resistance (sec)	135	135	135	135	135	135
Nitric Acid Resistance	Good	Good	Good	Good	Good	Good
Weld Characteristics Tensile Strength (kg/cm ²)	120	110	145	100	135	130
Appearance	Bad	Bad	Bad	Bad	Bad	Bad
Blooming Resistance (100°C; 240 hrs)	Good	Bad	Bad	Bad	Good	Good
Flame Retardance (UL94; Vertical Burning Test; 1.6t; Class)	*	V-2	*	V-2	V-2	*

* It did not fall under any class.

Table 4 (IV)

	<u>7</u>	<u>8</u>	<u>Comparative Example</u>			<u>11</u>	<u>12</u>
			<u>9</u>	<u>10</u>			
Tensile Strength (kg/cm ²)	430	400	300	300	280	600	
Heat Distortion Temp. (18.6 kg/cm ² ;°C)	130	120	90	100	90	140	
Arc Resistance (sec)	135	135	135	135	135	80	
Nitric Acid Resistance	Good	Good	Good	Good	Good	Good	
Weld Characteristics Tensile Strength (kg/cm ²)	130	120	180	180	170	150	
Appearance	Bad	Bad	Good	Good	Good	Good	
Blooming Resistance (100°C; 240 hrs)	Bad	Good	Good	Bad	Good	Good	
Flame Retardance (UL94;Vertical Burning Test; 1.6t;Class)	V-2	V-2	*	V-2	V-2	*	

Table 5 (I)

Comparative Example

Blending Materials (wt.%)

	13	14	15	16	17	18
PP Homopolymer	0	0	59	44	0	39
PP Block Copolymer	34	44	0	0	44	0
Glass Fiber/Modified PP	15/6	15/6	5/6	5/6	5/6	5/6
Talc/Mica	10/10	10/0	10/0	10/0	10/0	10/0
Silica	10	5	0	0	0	0
Calcium Carbonate	0	0	20	20	20	20
DBDE	10	15	0	10	10	15
Antimony Trioxide	5	5	0	5	5	5
PP Homopolymer:	Propylene homopolymer resin having a melt flow rate of 10g/10 minutes					
PP Block Copolymer:	Propylene-ethylene block copolymer resin having a melt flow rate of 10g/minutes and containing 8.5 wt.% of ethylene.					
Modified PP:	Maleic anhydride-modified propylene-ethylene copolymer resin.					
DBDE:	Decabromodiphenyl ether.					

Table 5 (II)

	13	14	Comparative Example			17	18
			15	16			
Tensile Strength (kg/cm ²)	550	500	400	370		330	320
Heat Distortion Temp. (18.6 kg/cm ² ; °C)	140	140	105	110		100	110
Arc Resistance (sec)	70	70	135	135		135	135
Nitric Acid Resistance	Good	Good	Bad	Bad		Bad	Bad
Weld Characteristics Tensile Strength (kg/cm ²)	138	150	180	165		150	145
Appearance	Good	Good	Good	Good		Good	Good
Blooming Resistance (100°C; 240 hrs)	Good	Good	Good	Bad		Good	Bad
Flame Retardance (UL94; Vertical Burning Test; 1.6t; Class)	V-2	V-1	*	*		*	V-2

* It did not fall under any class.

Claims

1. A resin composition for electric insulation materials for vehicles which is prepared by blending a crystalline propylene-ethylene copolymer resin with 3 to 10 wt.% of glass fibers, 10 to 20 wt.% of talc or mica, 5 to 20 wt.% of silica, 4 to 20 wt.% of a modified polyolefin resin, 10 to 30 wt.% of a halogen-containing flame retardant and 3 to 15 wt.% of a flame retardant auxiliary, said percentages being based on the weight of said resin composition.
2. A resin composition for electric insulation materials for vehicles according to Claim 1 wherein said crystalline propylene-ethylene copolymer resin contains 2 to 35 wt.% of ethylene and has a melt flow rate (MFR; a discharge of melted resin for 10 minutes in the case that 2.16 kg is loaded at a temperature of 230°C) of 1.0 to 80 g/10 minutes.
3. A resin composition for electric insulation materials for vehicles according to Claim 1 wherein an amount of said glass fibers is within the range of 5 to 8 wt.%.
4. A resin composition for electric insulation materials for vehicles according to Claim 1 wherein an amount of said silica is within the range of 5 to 15 wt.%.
5. A resin composition for electric insulation materials for vehicles according to Claim 1 wherein said modified polyolefin resin is that which is obtained by melting and kneading one or more polyolefin resins selected from a polyethylene, a polypropylene, a propylene-ethylene copolymer and a polybutene together with an unsaturated carboxylic acid or its anhydride in the presence of an organic peroxide.
6. A resin composition for electric insulation materials for vehicles according to Claim 1 wherein said halogen-containing flame retardant has a melting point of 200°C or more.
7. A resin composition for electric insulation materials for vehicles according to Claim 1 wherein said halogen-containing flame retardant is one selected from decabromodiphenyl ether, dodecachlorododecahydridimethanodibenzocyclooctene and mixtures thereof.
8. A resin composition for electric insulation materials for vehicles according to Claim 1 wherein and amount of said halogen-containing flame retardant is within the range of 10 to 20 wt.%.
9. A resin composition for electric insulation materials for vehicles according to Claim 1 wherein said flame retardant auxiliary comprises one or more of antimony compounds and boron compounds.
10. A resin composition for electric insulation materials for vehicles according to Claim 1 wherein said resin composition contains one or more of colorants such as dyes or pigments, nucleating agents, lubricants, anti-oxidants, heat stabilizers, light stabilizers, release agents, crosslinking auxiliaries, radical generators and foaming agents, as optional components.
11. A resin composition for distributor caps which is prepared by blending a crystalline propylene-ethylene copolymer resin with 3 to 10 wt.% of glass fibers, 10 to 20 wt.% of talc or mica, 5 to 20 wt.% of silica, 4 to 20 wt.% of a modified polyolefin resin, 10 to 30 wt.% of a halogen-containing flame retardant and 3 to 15 wt.% of a flame retardant auxiliary, said percentages being based on the weight of said resin composition.
12. A resin composition for distributor caps according to Claim 11 wherein said crystalline propylene-ethylene copolymer resin contains 2 to 35 wt.% of ethylene and has a melt flow rate (MFR; a discharge of a melted resin for 10 minutes in the case that 2.16 kg is loaded at a temperature of 230°C) of 1.0 to 80g/10 minutes.
13. A resin composition for distributor caps according to Claim 11 wherein an amount of said glass fiber is within the range of 5 to 8 wt.%.
14. A resin composition for distributor caps according to Claim 11 wherein an amount of said silica is within the range of 5 to 15 wt.%.
15. A resin composition for distributor caps according to Claim 11 wherein said modified polyolefin resin is that which is obtained by melting and kneading one or more polyolefin resins selected from a polyethylene, a polypropylene, a propylene-ethylene copolymer and a polybutene together with an unsaturated carboxylic acid or its anhydride in the presence of an organic peroxide.
16. A resin composition for distributor caps according to Claim 11 wherein said halogen-containing flame retardant has a melting point of 200°C or more.
17. A resin composition for distributor caps according to Claim 11 wherein said halogen-containing flame retardant is one selected from decabromodiphenyl ether, dodecachlorododecahydridimethanodibenzocyclooctene and mixture thereof.
18. A resin composition for distributor caps according to Claim 11 wherein an amount of said halogen-containing flame retardant is within the range of 10 to 20 wt.%.
19. A resin composition for distributor caps according to Claim 11 wherein said flame retardant auxiliary comprises one or more of antimony compounds and boron compounds.

20. A resin composition for distributor caps according to Claim 11 wherein said resin composition contains one or more of colorants such as dyes or pigments, nucleating agents, lubricants, anti-oxidants, heat stabilizers, light stabilizers, release agents, crosslinking auxiliaries, radical generators and foaming agents, as optional components.

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DOCUMENTS CONSIDERED TO BE RELEVANT			EP 87118549.2
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl. 4)
Y	<u>GB - A - 2 143 831</u> (MITSUBISHI) * Claims; page 4, line 118 - page 5, line 43; example 1 * --	1-3,5- 7,9-13, 15-17, 19,20	H 01 B 3/44 C 08 K 13/04 H 01 R 39/60
Y	CHEMICAL ABSTRACTS, vol. 93, no. 22, December 1, 1980, Columbus, Ohio, USA JAPAN ATOMIC ENERGY RESEARCH INSTITUTE FURUKAWA ELECTRIC CO LTD.; "Fire-resistant electric insulators" page 70 * abstract-no. 205 979t * & Jpn. Kokai Tokkyo Koho 80-81 419 --	1-3,5- 7,9-13, 15-17, 19,20	
A	<u>EP - A1 - 0 181 832</u> (CIBA-GEIGY) * Pages 1,7 * -----	1,11	TECHNICAL FIELDS SEARCHED (Int. Cl. 4) H 01 B 3/00 C 08 K H 01 R 39/00 C 08 L 23/00 F 02 P 7/00 G 12 B 9/00 H 05 K 5/00 H 02 K 5/00
The present search report has been drawn up for all claims			
Place of search VIENNA		Date of completion of the search 24-02-1988	Examiner KUTZELNIGG
CATEGORY OF CITED DOCUMENTS X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document	